



## Volatility of indoor and outdoor ultrafine particulate matter near a freeway

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### Abstract

Although recent studies have shown a positive association of exposure to ultrafine particulate matter (PM) with adverse effects on human health, it is not yet clear which PM components or properties of these particles may cause these responses. In the context of human exposure, depending on ventilation and air exchange ratios and in the absence of major indoor sources, an appreciable fraction of the indoor ultrafine aerosol is of outdoor origin. This study examined volatility of penetrating ultrafine outdoor particles, predominantly from freeway emissions, into indoor environments where other particle sources were minimized and no cooking activities took place. A tandem differential mobility analyzer (TDMA) system was used to study particle volatility at two apartments, 15 and 40 m downwind of the I-405 Freeway in Los Angeles, CA. The first differential mobility analyzer (DMA) selected particles of a certain diameter and subsequent heating of this monodisperse aerosol allowed for detection of changes in particle diameters by measuring the resulting size distribution with a second DMA. Aerosol volatility was examined by measuring changes in particle diameters as well as volume and number concentrations. Results suggest that outdoor particles are more volatile than indoor aerosols. Increasing temperature from ambient to 130 °C decreased and broadened indoor and outdoor aerosol mode diameters, however greater mode decreases were observed for outdoor particles. Furthermore, outdoor particles lost more of their volume upon heating than

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indoor aerosols. No significant particle losses due to volatilization were observed at 60 °C for either indoor or outdoor aerosols. A greater number of outdoor than indoor particles was lost at 110 °C. Heated outdoor particles with diameters greater than 45 nm showed bi-modal distributions, indicating that some of the aerosol is composed of primarily non-volatile particles, whereas the remaining particles are composed of mainly volatile material and consequently shrink. Evaluation of outdoor particle volatility as a function of distance to the freeway revealed that aerosol volatility decreases with increasing distance from the source.

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**Keywords:** Ultrafine particles; Indoor environments; Freeways; Volatility; Tandem differential mobility analyzer (TDMA)

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## 1. Introduction

Epidemiological studies have shown a positive association of exposure to ultrafine particulate matter (PM) with effects on human health (e.g. Donaldson et al., 2002; Li et al., 2003; MacNee and Donaldson, 2003). It is, however, not yet clear which chemical particle components or physical-morphological properties cause these adverse effects (Harrison and Yin, 2000).

Elevated ultrafine particle concentrations have been reported near major highways (Zhu et al., 2002), where many urban residents live. These residents may be exposed to potentially harmful ultrafine particles. Because people spend most of their time indoors (Jenkins et al., 1992), it is of interest to study transformation of outdoor aerosols after they penetrate into buildings. Particle volatility must be considered when evaluating penetration of outdoor particles indoors. Volatile aerosols of outdoor origin may decrease in size or evaporate entirely as they penetrate into indoor environments. During the colder period of the year, the indoor environment is typically warmer than the outdoors, therefore partial evaporation may occur slowly as the aerosol reaches indoor temperatures. However, heating is not the only way volatile material is evaporated from ultrafine PM. Dilution of volatile gases at increased distance from the freeway will cause the smaller particles in the ultrafine size range to shrink by evaporation (Zhang et al., 2004). Dilution of volatile material in the gas-phase caused by adsorption in buildings (Singer et al., 2004) may play a major role in evaporation from particles indoors. The smaller the particles the faster the evaporation due to the Kelvin effect. The resulting physical and chemical changes of outdoor particles will likely affect their toxicity and are important metrics in the development of fundamental understanding of the toxicity of PM originating from vehicular emissions. Recent emissions testing in either dynamometer (Sakurai et al., 2003a; Sakurai et al., 2003b) or on-road testing facilities (Kittelson et al., 2004) has shown that, depending on vehicle type, age and ambient conditions, over 90% of the particles by number and 10–30% by mass consist of more volatile material than others (known as semi-volatile), and upon heating, will partially evaporate.

For a better understanding of semi-volatile behavior of ultrafine PM a tandem differential mobility analyzer (TDMA) system can be used (Rader and McMurry, 1986). The first differential mobility analyzer (DMA) is used to select particles of a certain mobility diameter. Subsequent heating of these monodisperse particles allows for detection of changes in particle diameters by measuring the resulting size distribution with a second DMA. For example, TDMA systems have been used to measure the volatile sulfuric acid fraction of marine boundary layer aerosols (Orsini et al., 1999). These systems also measured the non-volatile carbonaceous fraction of particles in differently polluted urban and rural

environments (Philippin et al., 2004). In addition, TDMAs have been used to determine the mixing state of volatile and non-volatile components in carcinogenic diesel exhaust particles in dynamometer studies (Sakurai et al., 2003a).

This study was carried out together with experiments presented by Zhu et al. (2004), which focused on ultrafine particle size distribution and penetration efficiencies into indoor environments. In this study, we analyzed the volatility properties of freeway-generated particles. Measurements of the volatile fraction of both outdoor and indoor aerosols were performed using a TDMA in apartments close to the I-405 freeway in Los Angeles, CA. This study allowed for the examination of the external and internal mixing of volatile and non-volatile components of ultrafine freeway aerosol, and the determination of how aerosol particles change when they penetrate into buildings.

## 2. Methods

### 2.1. Tandem DMA system

The TDMA system used to measure volatility of particles is shown in Fig. 1. The system was operated inside an apartment. The two Scanning Mobility Particle Sizers (SMPS, TSI model 3936) consisted of a bipolar charger (with an  $^{85}\text{Kr}$  source), a long DMA (model 3081), and a Condensation Particle Counter (CPC, TSI model 3022A). The first SMPS (SMPS1) sampled either from outside the apartment or from inside. SMPS1 was set to select particles of a certain narrow size range. The resulting monodisperse aerosol was split into two streams. One stream went into the CPC of SMPS1 (control CPC, sample flow rate of  $0.31\text{ l min}^{-1}$ ), which measured the concentration of this monodisperse aerosol. The other stream traveled through a heater unit. After passing through the heater, the conditioned aerosol went through the second SMPS (SMPS2), which measured its size distribution. SMPS2 was used with a CPC sample flow rate of  $1.51\text{ l min}^{-1}$  and a sheath air flow rate of  $151\text{ l min}^{-1}$ .

The heater used to condition aerosols consisted of a 50 cm, straight stainless-steel tube with inside and outside diameters of 5.4 and 6.4 mm, respectively. Part of the tube (40 cm) was wrapped with a 125 W rope heater and insulated with a fiberglass blanket around the tube and the rope heater. Heater dimensions were similar to the system described by Orsini et al. (1999). Heater flow rate of  $1.51\text{ l min}^{-1}$  resulted in a Reynolds number of 380, indicating laminar flow conditions in the tube, and, therefore avoiding increased deposition losses associated with turbulent flow. The residence time of particles in the heated section of the tube was approximately 0.5 s, which is sufficient for volatilization to occur (Orsini et al., 1999).

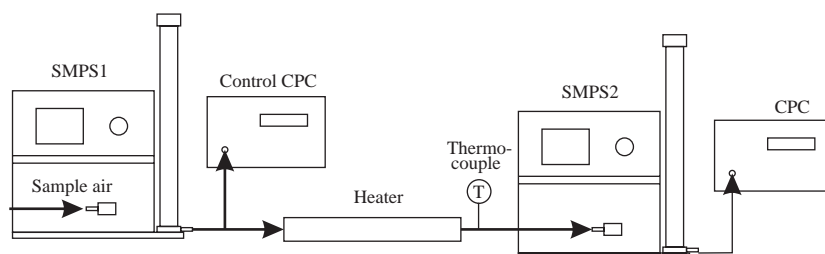


Fig. 1. Tandem DMA set-up.

The heater was thermally insulated by means of polytetrafluoroethylene (PTFE) connectors to upstream and downstream stainless-steel tubes. While the upstream tube was only 10 cm long, the length of the downstream tube was 30 cm, which allowed cooling of the aerosol before its introduction into SMPS2. The total length of this heater assembly was 0.91 m.

The heater was operated at AC voltages between 0 and 50 V (using a variable-output transformer) depending on the desired aerosol temperature inside the heater. The aerosol temperature was measured using a thermocouple probe at the center of the flow in the tube, introduced through a hole in the side of the tube just downstream of the heated section. The probe was fixed with epoxy glue and additionally sealed with silicone. A small thermocouple with wire diameters of 0.13 mm (Type K) was used to avoid flow obstruction.

Concerns may arise regarding re-condensation or re-nucleation of volatilized vapors when the aerosol exits the heated section and cools down to ambient temperature. However, when the aerosol is introduced in the heater the actual vapor pressure of the volatile material (in gas phase) is lower as compared to ambient conditions. This is because the DMA selects only particles of a certain size and vapors will only reach the heater by diffusion. Therefore, even after cooling back to ambient temperature, the vapor pressure in the gas surrounding the particles will be lower than the equilibrium vapor pressure of particles with volatile material. Under these conditions, the Kelvin effect does not allow re-condensation on the particles, which have the selected or, if shrunk, a smaller size. Sakurai et al. (2003a) did not observe re-condensation or re-nucleation when they measured volatility of diesel exhaust particles, with number concentrations higher than the concentrations observed during this study, using a similar TDMA design. Hence, re-condensation or re-nucleation was not expected in this study.

The Aerosol Instrument Manager software (version 5.2, TSI Inc., St. Paul, MN) was used for inversion of the measured CPC number concentrations to the size distribution. It was assumed that particles did not lose their charge during heating and evaporation of the volatile fraction, therefore they were still charged (as by electrostatic size selection by SMPS1) when entering SMPS2. However, some charge losses might occur before the particles are classified by SMPS2. To account for changes of particle charges and other size dependent losses, the selected monodisperse aerosol size distribution measured by SMPS2 at ambient temperature (heater switched off) was integrated to give the total particle number concentration. These values were compared to the concentration measured by the control CPC. Values from SMPS2 for diameters between 18 and 90 nm were generally between 40% and 65% of the control CPC concentrations. These size dependent ratios were then used to correct SMPS2 size distributions for heated and unheated aerosols.

## 2.2. *Sample locations and sample schedule*

Two two-bedroom apartments, in the vicinity of the I-405 Freeway in Los Angeles, CA were used for this study. These apartments are in identical buildings with the same interior layouts. The two apartments (Apt. 1 and Apt. 2) are on the eastern side of the I-405, and both are on the third floor with windows 3 m above a sound barrier wall. The distances between the wall and Apt. 1 and the wall and Apt. 2 are 15 and 40 m, respectively. The two apartments are separated by approximately 50 m. Both apartments are about 8 years old with central mechanical ventilation systems that was turned off during all measurements of outdoor and indoor aerosol volatility in this study.

I-405 runs generally north and south next to the apartment buildings. It has ten lanes near the sampling location, five northbound and five southbound, and it is about 40 m wide with a 1 m wide median strip

and two 2 m wide shoulders. The freeway is on the same level as the surrounding terrain of the sampling site. The apartments lie approximately 6 km to the east of Santa Monica Bay and the Pacific Ocean. More details of the sampling sites and the meteorological conditions are described in a related work (Zhu et al., 2004).

The study took place from December 2003 to January 2004. Measurements in the two apartments were performed during the day between 10 am and 7 pm. Apt. 1 was sampled for 11 days between December 19, 2003 and January 6, 2004, and measurements in Apt. 2 were completed in 6 days, between January 7 and 12, 2004.

### 2.3. Measurements

The TDMA system was used to sample particles of a fixed diameter at a constant aerosol temperature. Four to eight SMPS2 scans of 2 min length each were used to obtain average size distributions of the conditioned aerosol. Concurrently, the control CPC measured average number concentrations of the aerosol selected by SMPS1 with 2-min averaging times. First, measurements at ambient temperature were performed for aerosols of various diameters, sampled consecutively from outdoor and indoor air. This was followed by outdoor and indoor measurements of the same particle sizes at one heated aerosol temperature. This sequence of measurements lasted approximately 3 h, and was repeated three times during the sampling period. This sampling protocol was repeated for an additional aerosol temperature. In Apt. 1, the particle sizes used for the sampling protocol were 18, 45, and 90 nm. In addition, 27 nm particles were sampled using a modified protocol, which used two heated aerosol temperatures within the same sequence of measurements.

For heating, the aerosol temperatures of 60 and 110 °C were chosen to determine ultrafine particle volatility. We expected most of the volatile material to evaporate at 110 °C (Philippin et al., 2004). The temperature of 60 °C was selected to represent an intermediate state of volatilization. Additional temperatures of 90 and 130 °C were used to create a thermal desorption profile of outdoor aerosol in the proximity of freeways. Several scans were repeated by DMA2 at each of the temperatures to measure the temperature dependence of a selected particle size.

## 3. Results and discussion

Figs. 2 and 3 show typical results from measurements of outdoor particles in Apt. 1 (15 m from freeway). Each figure shows three normalized number size distributions: the first represents the selected monodisperse aerosol without conditioning (no heating) at ambient temperature; the second shows the size distribution of the original monodisperse aerosol after heating to 60 °C; the third shows the size distribution after heating to 110 °C. The size distributions were initially normalized using the measured concentration of the control CPC to account for changes in the ambient particle concentrations. In addition, the distributions were normalized so that the size distribution of the monodisperse aerosol without conditioning exhibited a maximum at 1. As expected, these measurements show that the mode diameter  $d_m$  of the originally monodisperse aerosol decreases towards a smaller diameter with increasing heater temperature (mode decrease). The mode also broadens with increasing temperature because not all particles shrink to the same degree due to differences in their chemical composition.

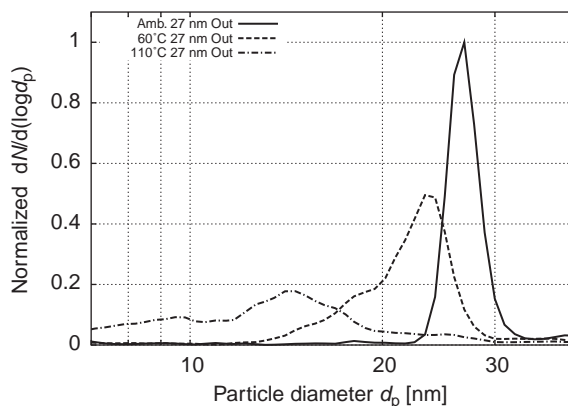


Fig. 2. Normalized number size distribution of 27 nm outdoor particles at a distance of 15 m from the I-405 freeway at ambient temperature ( $\approx 25^\circ\text{C}$ ) and after conditioning (heating) at 60 and  $110^\circ\text{C}$ .

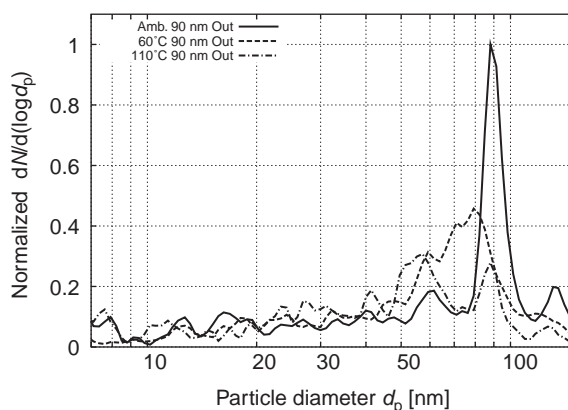


Fig. 3. Normalized number size distribution of 90 nm outdoor particles at a distance of 15 m from the I-405 freeway at ambient temperature ( $\approx 25^\circ\text{C}$ ) and after conditioning (heating) at 60 and  $110^\circ\text{C}$ .

The size distribution of ambient 90 nm particles depicts two smaller peaks to the right and the left side of the major mode. These peaks are caused by the increased probability of doubly charged particles at this diameter (Wiedensohler, 1988), so that some of the particles selected by SMPS1 had a greater size but carried two elementary charges (resulting in the same electrical mobility). When these particles change their charge from two to one they are measured as greater particles by SMPS2. Particles that had one elementary charge in SMPS1 and changed to two charges in SMPS2 are measured as smaller particles. The effect of doubly charged particles was not considered in this work, as they became apparent only in case of the 90 nm particles, where the effect was still small enough not to contribute significantly to the modes observed after heating.

The 90 nm aerosol size distribution split into two modes at heater temperatures of  $90^\circ\text{C}$  and above, as can be seen in Fig. 3 for the curve representing a heater temperature of  $110^\circ\text{C}$ . One mode remains close to the monodisperse mode of the unheated aerosol, while the other mode is at a lower diameter.

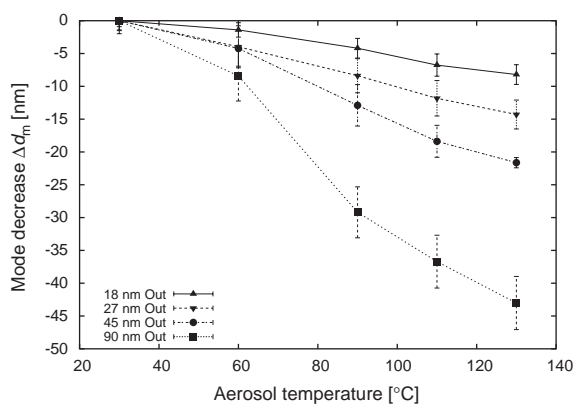


Fig. 4. Mode diameter decrease  $\Delta d_m$  as a function of aerosol conditioning temperature. The error bars for the 45 and 90 nm particles are standard deviations from four measurements on different days. For 18 and 27 nm particles, only two and one measurements, respectively, were performed, hence the error bars for these two sizes represent modes widths (at half-amplitude).

This bi-modal distribution indicates that a fraction of the 90 nm aerosol consists of particles that are composed of almost entirely non-volatile material. This part of the aerosol contributes to the mode close to the original mode diameter at ambient temperature, not broadened significantly (non-volatile mode). The particles of the remaining portion contain mostly volatile material and continue to shrink in size with increasing temperature; they contribute to the mode at smaller diameter, which is significantly broadened (mostly volatile mode).

### 3.1. Temperature dependence

To study temperature dependence of the mode decrease more in detail, for particles with diameters of 18, 27, 45, and 90 nm the mode diameter was determined from measurements at aerosol conditioning temperatures of 30, 60, 90, 110, and 130 °C. The particle size to be analyzed by the TDMA was set manually at SMPS1 and differed from the mode diameter of this monodisperse aerosol measured by SMPS2 at ambient temperature. After considering temperature and pressure conditions for both SMPS, the aerosol size measured by SMPS2 was approximately 9% lower than the size set for SMPS1. The reason for this difference is not clear, and, in part, it might be due to evaporation of highly volatile material from the particles between SMPS1 and SMPS2 at ambient temperature. However, it is not believed that this would be responsible for a 9% decrease in particle diameter, independently from the particle size being analyzed. Hence, for all results presented in this study, the size of the monodisperse aerosol is reported as the mode diameter measured by SMPS2 at ambient temperature. Therefore, the mode decrease  $\Delta d_m$  is 0 nm at 30 °C, considered as ambient temperature for this measurement.

Fig. 4 shows the mode decrease  $\Delta d_m$  as a function of aerosol temperature resulting from these measurements (thermal desorption profile), where  $\Delta d_m$  is the difference of the measured mode diameter to the selected particle diameter. In case of bi-modal distributions, the mode decrease of the mostly volatile mode is shown in Fig. 4. The trend observed in this figure shows that for temperatures of up to 130 °C the diameter of all particles studied (18–90 nm) is still shrinking without a clear evidence of a plateau indicating the presence of a non-volatile core. If a non-volatile core exists, its diameter will be smaller

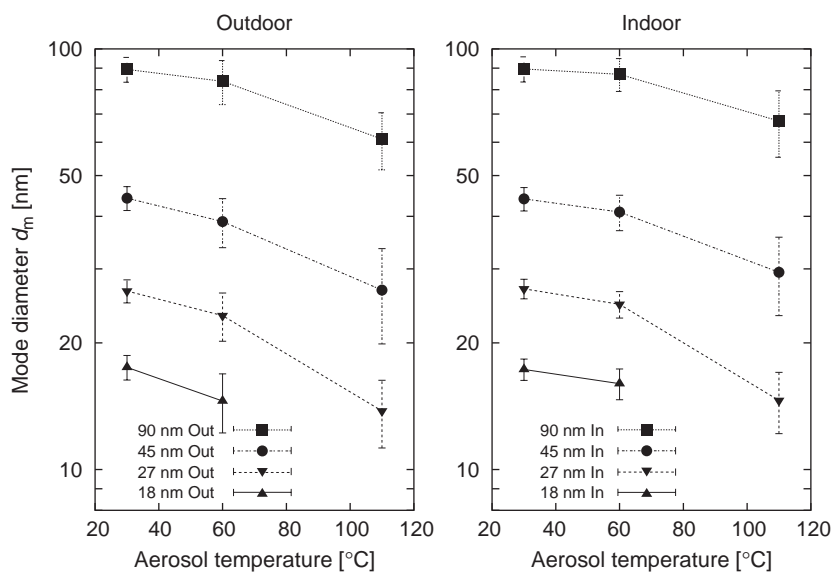


Fig. 5. Mode diameters  $d_m$  as a function of temperature for 18, 27, 45, and 90 nm Apt. 1 outdoor and indoor particles. Averages were obtained based on three or four measurements in most cases (only two measurements for 45 nm indoor and outdoor and 27 nm indoor particles heated to 110 °C). Error bars indicate mode widths at half-amplitude of the average size distribution, demonstrating mode broadening with increasing temperature.

than that reached by particles at 130 °C (approximately half the diameter of the respective aerosols at ambient temperature). Comparison of these results with the mode decrease reported by Sakurai et al. (2003b) for diesel aerosol measured on a dynamometer, reveals a similar profile. However, the mode diameters decreased more for diesel aerosol. Sakurai et al. (2003b) observed a leveling off after about 110 °C, suggesting the existence of a non-volatile core for diesel aerosol.

### 3.2. Comparison of outdoor and indoor volatility

Comparison of particle size distributions of heated outdoor aerosol with size distributions of heated indoor aerosol reveals that outdoor particles shrink more than indoor particles. This result suggests that outdoor particles are more volatile than indoor particles, which may be caused by evaporation of some of the volatile material from the outdoor particles as they penetrate into indoor environments. Lunden et al. (2003) observed losses of nitrate from particles penetrating a building, which they explained by dissociation and evaporation and subsequent losses by deposition and sorption to indoor surfaces.

Fig. 5 illustrates that outdoor particles are more volatile than indoor particles for all studied diameters. The difference between outdoor and indoor mode decreases is statistically significant at 60 °C for all particle sizes and at 110 °C for 90 nm particles. The results at 110 °C seem to be less clear, and are attributed to less distinct modes at this temperature and the difficulties in determining mode diameters from the measurements.

The particle number size distributions can be integrated to yield total number concentrations. By comparing the total number concentrations of heated aerosols to the total number concentration of the

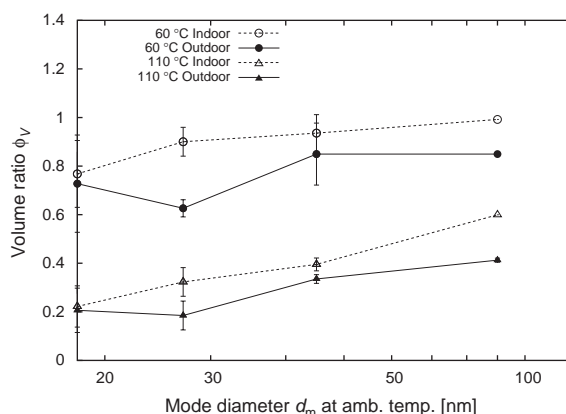


Fig. 6. Comparison of fractions  $\phi_V$  for outdoor and indoor aerosols at Apt. 1.

aerosol at ambient temperature, one could determine whether heating results in particle losses. Normalized total number concentrations were obtained by integrating the SMPS size distributions and dividing by the control CPC average concentrations, and to account for changes in particle number concentration of the sampled aerosol during the measurements. To evaluate the losses, we defined the parameter  $\phi_N$  as the ratio of the normalized total number concentrations for heated aerosols to the normalized total number concentrations of the aerosol at ambient temperature. Similarly, a volume-based parameter,  $\phi_V$ , was defined as the ratio of the normalized total volume concentrations for heated aerosols to the normalized total volume concentrations of the aerosol at ambient temperature. The ratios  $\phi_N$  and  $\phi_V$  thus represent the fraction of the aerosol not lost due to volatilization at a given temperature in terms of number concentration and volume, respectively.

In most cases, heating did not result in significant particle number losses. Only 18 nm particles heated to 110 °C were lower in number concentrations than those at ambient temperature: both indoor and outdoor particles lost between 50% and 60%. This may be due to complete particle volatilization or shrinkage to sizes below the CPCs detection limit of 7 nm. Outdoor aerosols of other sizes experienced minor number losses of about 20%.

Particle volume, used to determine the fraction of volatilized material, was estimated assuming spherical particles. As indicated by the fractions  $\phi_V$  shown in Fig. 6, volume losses increase significantly from 60 to 110 °C, and are generally higher for outdoor than indoor particles for all sizes tested. This observation is consistent with the notion that outdoor particles appear to be more volatile than indoor particles. Smaller particles are also shown to lose more of their volume, indicating that they may be composed of less non-volatile material.

The bi-modal distribution observed for 90 nm particles (shown in Fig. 3) and also for 45 nm particles indicated external mixing of non-volatile and mostly volatile particles. Integration of the number size distribution over the size range occupied by the non-volatile mode yields the number concentration of non-volatile particles. The ratio of this number concentration to the number concentration of the original mode (integration of distribution at ambient temperature) represents the fraction of non-volatile particles in the aerosol. For the 90 nm outdoor aerosol (shown in Fig. 3) the ratio is 15%, while it is 18% for indoor 90 nm particles. For 45 nm particles, the number fractions of non-volatile

Table 1

Outdoor particle mode decreases compared for Apt. 1 and Apt. 2 at 60 and 110 °C.

$d_p$ (nm)	$\Delta d_m$ at Apt. 1 (nm)		$\Delta d_m$ at Apt. 2 (nm)	
	60 °C	110 °C	60 °C	110 °C
18	$-3.0 \pm 0.6$	—	$-3.1 \pm 0.9$	–8.7
27	$-3.3 \pm 0.7$	$-12.8 \pm 1.9$	$-4.0 \pm 1.6$	—
45	$-5.6 \pm 2.6$	$-17.0 \pm 1.3$	$-3.4 \pm 0.4$	–15.9
90	$-6.1 \pm 5.8$	$-27.7 \pm 5.9$	$-4.0 \pm 1.0$	$-27.2 \pm 6.0$

outdoor and indoor particles are 17% and 15%, respectively. The 18 and 27 nm aerosols each showed only one mode with its diameter shrinking with increasing temperature. This suggests that particles of these sizes were, in terms of volatility, only internally mixed and predominantly composed of volatile material.

The outdoor and indoor aerosol seem to have similar external mixing ratios. If size changes occur in the mostly volatile part of the aerosol while it penetrates the building, it appears that they do not change the concentration of these particles in relation to the non-volatile particles. Particles shrinking from 45 or 90 nm to a smaller size may be replaced by a similar amount of originally larger outdoor particles that shrink to 45 or 90 nm indoor particles. The external mixing observed for 45 and 90 nm particles may result from diesel exhaust. On a dynamometer, 70 nm diesel exhaust particles have been shown to produce a bi-modal distribution (Sakurai et al., 2003a) as well as 150 nm particles in a highly polluted environment (Philippin et al., 2004).

### 3.3. Dependence on distance from freeway

Outdoor particle size distributions of ambient and heated aerosols were also measured in Apt. 2, 40 m downwind of the freeway to evaluate changes of the volatility properties at a greater distance from the freeway. At Apt. 2 the size distribution of aerosol at 110 °C shows a mostly volatile mode, which is less distinct and broader than at Apt. 1, 15 m downwind of the freeway. This implies that particles further from the source may experience more mixing with background aerosol having different volatilities.

Table 1 shows mode decreases  $\Delta d_m$  for both apartments. The mode decreases of outdoor aerosol at Apt. 2 seem to be similar for 18 and 27 nm aerosols and slightly lower for 45 and 90 nm aerosols when compared to mode decreases of outdoor aerosol at Apt. 1. This observation supports the argument that outdoor aerosol volatility is lower at increased distance from a freeway. This trend seems to be less pronounced at the smallest particle sizes of 18 and 27 nm.

A comparison of the values of  $\phi_N$  for the two apartments indicates insignificant (if any) particle number losses due to heating at both apartments at 60 °C. For 110 °C there are about 20% losses of 27 nm particles but no losses of 45 and 90 nm particles at Apt. 2, while at Apt. 1 27, 45, and 90 nm aerosols each lose approximately 20% in number. Losses of 18 nm particles are difficult to compare due to greater standard deviations. Volume losses are shown in Fig. 7 by means of the ratios  $\phi_V$  for both apartments. Results for 18 nm particles at Apt. 2 are not shown because of the large uncertainties in volume estimates due to low particle concentrations encountered. The results in Fig. 7 show the same decreasing trend in volume-based aerosol volatility at increasing distance from the freeway, thereby confirming the results obtained from

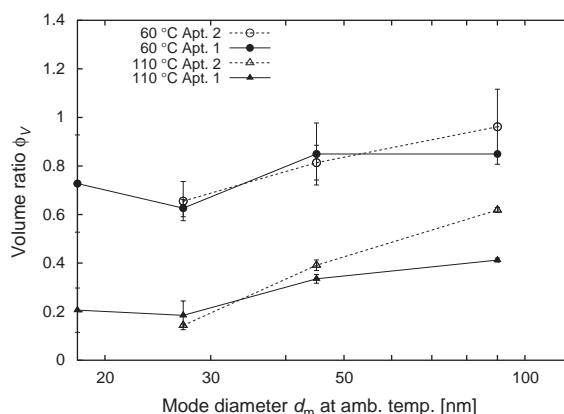


Fig. 7. Comparison of fractions  $\phi_V$  between Apt. 1 and Apt. 2 outdoor aerosols.

the mode decrease at the two apartments. Again, the decreasing volatility is more evident at the larger particle sizes.

#### 4. Summary and conclusions

Volatility properties of freeway ultrafine outdoor and indoor aerosols were examined by heating particles and detecting changes in their diameters and number concentrations. Results from this study suggest that outdoor particles are generally more volatile than indoor aerosols, for all studied particle sizes. Although the aerosol mode diameter broadened for both indoor and outdoor particles, increase in aerosol temperature resulted in greater decrease of mode diameter for outdoor particles. In addition, outdoor particles lost more of their volume upon heating than their indoor counterparts. Measurements of particle number concentrations revealed that more of outdoor than indoor particles were lost at 110 °C. At 60 °C heating did not result in significant losses in particle number for both indoor and outdoor aerosols.

Bi-modal distributions were observed for outdoor 90 and 45 nm particles heated to 110 °C. These distributions indicate that a fraction of the aerosol consists of non-volatile particles, whereas the other particles are mostly volatile and therefore shrink upon heating. No bi-modal distribution was observed for 18 and 27 nm particles, suggesting that all of these particles are composed mostly of volatile material.

Increasing temperature causes particles to shrink further. For instance, when heated to 130 °C, particles shrink to approximately half of their original size. A trend in the temperature dependence of the mode decrease indicates that a fraction of heated aerosol might reach a final particle size, a non-volatile core. Finally, outdoor sampling in a location further away from the freeway showed that aerosol volatility decreases with increasing distance from the source. The difference in volatility, however, is small and less evident for 18 and 27 nm particles.

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## References

- Donaldson, K., Brown, D., Clouter, A., Duffin, R., MacNee, W., Renwick, L., Tran, L., & Stone, V. (2002). The pulmonary toxicology of ultrafine particles. *Journal of Aerosol Medicine*, 15, 213–220.
- Harrison, R. M., & Yin, J. (2000). Particulate matter in the atmosphere: which particle properties are important for its effects on health. *The Science of the Total Environment*, 249, 85–101.
- Jenkins, P. L., Phillips, T. J., Mulberg, E. J., & Hui, S. P. (1992). Activity patterns of Californians: use of and proximity to indoor pollutant sources. *Atmospheric Environment, Part A: General Topics*, 26, 2141–2148.
- Kittelson, D. B., Watts, W. F., & Johnson, J. P. (2004). Nanoparticle emissions on Minnesota highways. *Atmospheric Environment*, 38, 9–19.
- Li, N., Sioutas, C., Cho, A., Schmitz, D., Misra, C., Sempf, J., Wang, M., Oberley, T., Froines, J., & Nel, A. (2003). Ultrafine particulate pollutants induce oxidative stress and mitochondrial damage. *Environmental Health Perspectives*, 111, 455–460.
- Lunden, M. M., Revzan, K. L., Fischer, M. L., Thatcher, T. L., Littlejohn, D., Hering, S. V., & Brown, N. J. (2003). The transformation of outdoor ammonium nitrate aerosols in the indoor environment. *Atmospheric Environment*, 37, 5633–5644.
- MacNee, W., & Donaldson, K. (2003). Mechanism of lung injury caused by PM10 and ultrafine particles with special reference to COPD. *European Respiratory Journal*, 21, 47s–51s.
- Orsini, D. A., Wiedensohler, A., Stratmann, F., & Covert, D. S. (1999). A new volatility tandem differential mobility analyzer to measure the volatile sulfuric acid aerosol fraction. *Journal of Atmospheric and Oceanic Technology*, 16, 760–772.
- Philippin, S., Wiedensohler, A., & Stratmann, F. (2004). Measurements of non-volatile fractions of pollution aerosols with an eight-tube volatility tandem differential mobility analyzer (VTDMA-8). *Journal of Aerosol Science*, 35, 185–203.
- Rader, D. J., & McMurry, P. H. (1986). Application of the tandem differential mobility analyzer to studies of droplet growth or evaporation. *Journal of Aerosol Science*, 17, 771–787.
- Sakurai, H., Park, K., McMurry, P. H., Zarling, D. D., Kittelson, D. B., & Ziemann, P. J. (2003a). Size-dependent mixing characteristics of volatile and nonvolatile components in diesel exhaust aerosols. *Environmental Science and Technology*, 37, 5487–5495.
- Sakurai, H., Tobias, H. J., Park, K., Zarling, D., Docherty, S., Kittelson, D. B., McMurry, P. H., & Ziemann, P. J. (2003b). On-line measurements of diesel nanoparticle composition and volatility. *Atmospheric Environment*, 37, 1199–1210.
- Singer, B. C., Revzan, K. L., Hotchi, T., Hodgson, A. T., & Brown, N. J. (2004). Sorption of organic gases in a furnished room. *Atmospheric Environment*, 38, 2483–2494.
- Wiedensohler, A. (1988). An approximation of bipolar charge distribution for particles in the submicron range, technical note. *Journal of Aerosol Science*, 19, 387–389.
- Zhang, K. M., Wexler, A. S., Zhu, Y. F., Hinds, W. C., & Sioutas, C. (2004). Evolution of particle number distribution near roadways, Part II: the ‘Road-to-Ambient’ process. *Atmospheric Environment*, 38, 6655–6665.
- Zhu, Y. F., Hinds, W. C., Kim, S., & Sioutas, C. (2002). Concentration and size distribution of ultrafine particles near a major highway. *Journal of the Air & Waste Management Association*, 52, 1032–1042.
- Zhu, Y., Hinds, W. C., Krudysz, M., Kuhn, T., Froines, J., & Sioutas, C. (2004). Penetration of freeway ultrafine particles into indoor environments. *Journal of Aerosol Science*, submitted for publication, doi:10.1016/j.aerosci.2004.09.007.